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## NUCLEAR MATERIALS ACCOUNTING, HELPING THE FACILITY OPERATOR\*

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### ABSTRACT

A modern materials control and accounting (MC&A) system can provide major benefits to production personnel. It can enhance understanding of process systems performance, localize and reconcile material losses, and identify instruments that are out-of-calibration or malfunctioning. Examples of the above MC&A system applications are given. We show how Operations personnel can use an MC&A system to their advantage rather than letting the MC&A system take advantage of them.

### I. SUMMARY

A materials control and accounting (MC&A) system can help operators follow process performance and detect nonstandard conditions. The MC&A system can pinpoint both the source and time of material loss. The frequent closure of materials balances on small unit process areas can minimize the magnitude of material losses and clearly show that these losses result from process upsets or an inability to precisely measure all material quantities rather than an unauthorized removal.

A modern MC&A system can also quantify material loss to streams where measurements are impractical, can identify process upsets or equipment misoperation, and can improve understanding of the variables impacting process performance. Operational benefits are realized through an increase in both plant efficiency and product output.

The current major impediment to reducing plant inventory differences (IDs) upon materials balance closure is "unmeasured inventory" (holdup) in process equipment and glove boxes. A properly designed MC&A system can help in the development of estimators that predict the buildup of "unmeasured inventory" and tell the operator if cleanout is warranted.

The primary purpose of this paper is to advocate a positive interaction between Safeguards

and Operations personnel as opposed to the adversarial relationships that we see in some facilities. Means by which this goal can be achieved and examples of how the safeguards system can produce positive benefits in plant operations are discussed and illustrated.

### II. INTRODUCTION

Nuclear materials accounting is an important component of the nuclear fuel cycle. It provides assurance to the physical protection component of Safeguards that nuclear material is present and at its proper location or in the event that this is not so, initiates appropriate responses. Materials accounting also provides information that is useful to Health and Safety personnel and to Operations personnel.

Materials accounting is frequently viewed as a hindrance rather than as a help by the Operations personnel with whom we must work. Materials accounting requires the investment of Operations manpower or capital that could be productively used elsewhere in the plant. MC&A can be an inconvenience and at worst it can result in curtailed production and loss of job.

Safeguards personnel are often viewed as adversaries rather than as a help. Because of these negative perceptions, Safeguards personnel are not always welcomed by Operations. We who are members of the materials accounting community must reverse this perception if we are to effectively discharge our responsibilities.

To gain a more productive and positive interaction with Operations, the materials accounting community must work cooperatively with production personnel and demonstrate that materials accounting can be helpful in meeting production objectives; that is, we must develop interactions that yield production benefits. To accomplish this objective, materials accounting personnel must work with Operations to minimize the perceived negative impact of the MC&A requirements.

Safeguards personnel should be involved at the first stages of process and facility design.

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Their primary thrust should be to provide an effective materials accounting system that provides process control and other operational benefits while limiting cost impacts. The MC&A system designer should attempt to provide net cost reductions to the operator by using the capabilities of the MC&A system for process monitoring and control.

MC&A should use process control measurements whenever feasible. Where accountability grade measurements are not available, special nuclear material (SNM) values may be inferred from secondary correlations using process measurements (density, historical values, etc.) or other attributes (volume transferred, etc.). We sometimes require installation of better measurement systems because they exist, when in reality they may not result in significant materials accounting improvements. Special measurements for materials accounting purposes should be imposed only where a clear need can be shown to exist and where cost effectiveness can be demonstrated. In the coming era of tight budgets, the concept of cost effectiveness will increase in importance.

The MC&A system should not be an impediment to plant operations and production. When used properly, a good MC&A system can provide an enhanced understanding of process systems performance. A good MC&A system can identify, localize, and explain apparent "material losses," thus relieving the pressures from external auditing organizations. This concept should be stressed when dealing with Operations personnel.

### III. ACCOUNTABILITY MEASUREMENTS AND VARIANCE DETERMINATION

An analysis of the proposed system should be made to determine where materials accounting measurements can be most effective and, more importantly to the operator, where they are not warranted and alternate procedures should be considered. To illustrate this point, we will examine a hypothetical process that is a combination of several processes commonly used in Department of Energy facilities.

The process (Fig. 1) converts SNM in aqueous solution to metal. This process includes three major processing steps:

- feed purification and concentration by ion exchange,
- conversion to solid by precipitation, and
- conversion of the precipitate to a metal by reduction.

The example process receives 100 kg of feed as 10 batches of nitrate solution, each containing 10 kg of SNM. The primary output consists of 80 SNM buttons weighing approximately 1.1 kg each. Other outputs are

- ion exchange column effluent,
- filtrate from the precipitate filtration step,
- sweepings from the process line, and
- scrap from the calcium reduction step.

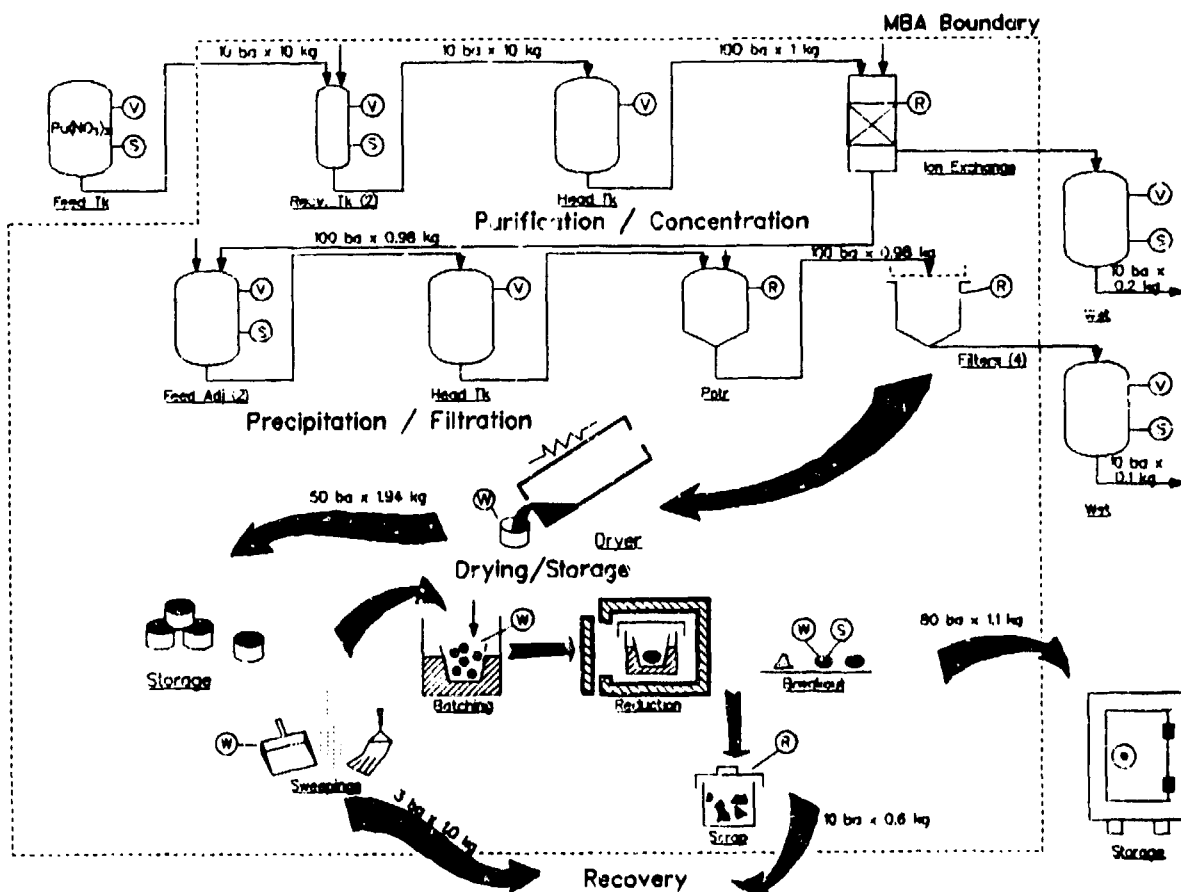
If accurate materials accounting is to be accomplished, measurements should be made to determine SNM content of all streams entering and leaving the materials balance area (MBA) and of all SNM inventories within the MBA. In some cases (spillage, holdup in transfer piping and complex geometry vessels, etc.), measurements may be impossible or at best quality measurements may be impractical. In these cases, it may be necessary to use estimators of unmeasured inventory (holdup) content if all terms in the materials balance equation are to be addressed.

Data on errors associated with each measurement or inventory estimate must also be available for variance calculations (error propagation). Tables I and II show the measurement values and errors associated with transfers and inventory determinations respectively for the example process. These errors are typical of those observed in operating facilities. Results of a variance calculation using the data from Tables I and II are summarized in Table III.

Table III tabulates the variance for individual inventories and transfers in the example process after 100 kg of feed has been processed. The uncertainty in the materials balance closure is 5.2 kg at one standard deviation. Review of these data shows that the inventory variances are the major contributors to uncertainty in the materials balance closure. The uncertainty on inventory in the dryer is the major contributor (18.0 kg<sup>2</sup>) to total inventory variance. Four of the process units provide inventory variance contributions of 1.0 kg<sup>2</sup> or greater. None of these variances approach that of the Dryer variance. At 100 kg throughput, the transfer terms do not make a significant contribution to total system variance.

The results of the above analysis indicate that for materials accounting purposes, the uncertainty on SNM inventory in the Dryer is the most significant contributor to total measurement uncertainty. The Dryer design should be examined to see if redesign could effect a reduction in Dryer holdup. The design should also be modified to facilitate Dryer cleanout to a low or constant level at the time of materials balance closure.

The SNM Storage can also contribute to significant uncertainty if SNM inventories are allowed to fluctuate at the time of materials balance closure. If the SNM inventory were to vary by 1.0 kg across the inventory period, the total variance would increase by 10 kg<sup>2</sup>. Addition of an SNM assay device to measure this material with a high precision, thus reducing storage variance, could be considered. This could require a major investment of capital to design and build the non-destructive assay (NDA) instrument and a major investment of manpower to operate it. It could be more cost effective to operate with minimum storage capacity or to run the storage down to zero or near-zero inventory before materials balance closure.



Legend: R-radiation, S-sample, (analytical), V-volume, W-weight

Fig. 1.  
The example process—nitrate-to-metal conversion.

TABLE I  
EXAMPLE PROCESS INVENTORY MEASUREMENTS AND ERRORS

Description	No.	Bulk Quantity	Percent Error		SNM Conc.	Percent Error	
			Random	System		Random	System
Feed Recvr.	2	2000 l.	5.0	3.0	0.005 kg/L	4.0	1.0
IX Head Tk.	1	2500 L	6.0	5.0	0.004 kg/L	4.0	1.0
IX Column	1	1.00 kg	5.0	25.0	1.0 kg/kg		
Feed Adj. Tk	2	400 L	6.0	4.0	0.025 kg/L	2.0	0.5
Prcep.Hd. Tk.	1	500 L	6.0	4.0	0.020 kg/L	2.0	0.5
Precipitator	1	2.00 kg	25.0	10.0	1.0 kg/kg		
Filters	4	1.25 kg	0.5	1.0	0.800 kg/kg	20.0	5.0
Dryer	1	10.0 kg	30.0				
NM Storage	10	1.11 kg	0.5	1.0	0.901 kg/kg	5.0	5.0
Reduction	2	2.61 kg	0.3	0.7	0.901 kg/kg	15.0	10.0
Breakout	1	2.35 kg	0.1	0.05	0.998 kg/kg	0.1	0.1
Sweepings	2	2.00 kg	0.4	0.8	0.500 kg/kg	50.0	
Scrap	3	0.90 kg	0.6	1.2	0.667 kg/kg	25.0	5.0

TABLE II  
EXAMPLE PROCESS TRANSFER MEASUREMENTS AND ERRORS

<u>Description</u>	<u>No.</u>	<u>Bulk* Quantity</u>	<u>Percent Error</u>		<u>SNM Conc.</u>	<u>Percent Error</u>	
			<u>Random</u>	<u>System</u>		<u>Random</u>	<u>System</u>
Feed	10	2000 L	0.5	0.3	0.005 kg/L	0.4	0.1
IX Waste	10	-2500 L	10.0	4.0	0.0001 kg/L	5.0	1.0
Filtrate	10	-500 L	10.0	4.0	0.0002 kg/L	5.0	1.0
Sweepings	3	-2.00 kg	0.4	0.8	0.500 kg/kg	50.0	
Scrap	10	-0.90 kg	0.6	1.2	0.667 kg/kg	25.0	5.0
Product	40	-2.20 kg	1.0	0.05	0.999 kg/kg	0.01	0.01

\*Positive = input; negative = output.

TABLE III  
EXAMPLE PROCESS VARIANCES

INVENTORY VARIANCES

<u>Description</u>	<u>Variance (kg<sup>2</sup>)</u>
Feed Receiver	1.64
IX Head Tank	1.60
IX Column	0.00
Feed Adjust Tank	1.60
Precipitator Head Tank	0.80
Precipitator	0.50
Filters	0.32
Dryer	18.00
SNM Storage	0.05
Reduction	0.50
Breakout	0.00
Sweepings	1.00
Scrap	0.14
Total Inventory Variance	26.15

TRANSFER VARIANCES

<u>Description</u>	<u>Variance (kg<sup>2</sup>)</u>
Feed	0.14
IX Waste	0.01
Filtrate	0.00
Sweepings	0.75
Scrap	0.32
Product	0.02
Total Transfer Variance	1.24
Total MBA Variance =	27.40 kg <sup>2</sup>
Total MBA Standard Deviation =	5.23 kg

The Dryer and the SNM Storage area are prime areas for diversion because detection sensitivity is poor in these areas relative to other parts of the process. Therefore, it would be prudent to invest more effort in physical protection for these areas. Limited access with close surveillance of personnel entering and leaving these areas should be considered as an adjunct or substitute for detailed materials accounting.

The Dryer and SNM Storage area might also be treated as a separate MBA. This would improve the loss detection sensitivity of the remaining MBAs located upstream and downstream. This assumes that good measurements can be made of transferred

Although improvements in the inventory measurement may not appear warranted, an alternate, cost-effective strategy should be considered. Draining of vessels containing solutions and cleanout of solids handling vessels at inventory will reduce the magnitude of these terms in the materials balance equation to effectively zero. This would be impractical if daily balances are attempted. However, for long inventory periods, for example, monthly, cleanout and flushdown will greatly reduce uncertainty.

Figure 2 shows the effect of throughput on uncertainty in the materials balance for the example process. The uncertainty is relatively constant at approximately 5.2 kg for throughputs below 200 kg. At throughputs above 500 kg, the transfer terms become important and they begin to dominate above a throughput of 1000 kg. The contributions of the inventory and transfer components are also shown in Fig. 2.

The contribution of individual transfers to total transfer uncertainty as a function of system throughput is shown in Fig. 3. Three transfers provide significant contributions to the total transfer variance. These are the large input feed stream and the two solid waste outputs (sweepings and scrap) that have small flows relative to the feed. For this particular system, the small throughput sweepings and scrap transfers contribute more to the total transfer variance than does

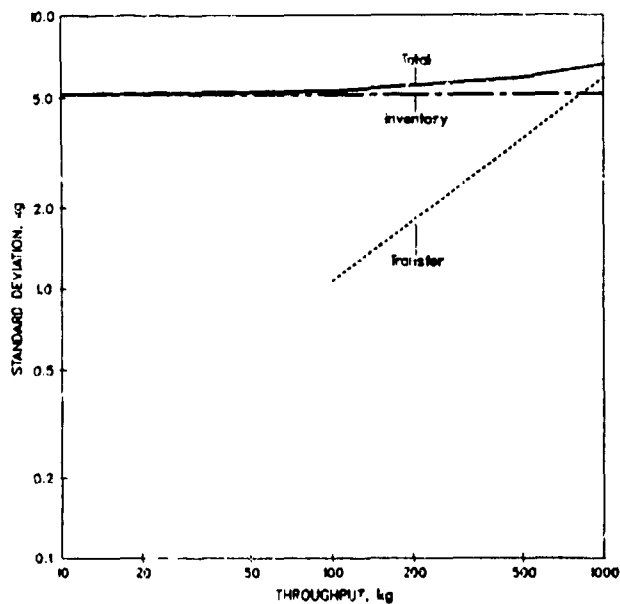


Fig. 2.

Materials balance uncertainty vs throughput.

the feed. For this particular case, it is apparent that improved measurement on sweepings and scrap could effect a significant reduction in the transfer variance. Although the measurement errors on ion exchange waste and filtrate are larger than for the feed, the variance contribution of these terms is relatively low because of the low transfer amount. An improvement in these measurements would be of no significant benefit.

The primary conclusions of the above exercise are the following.

- (1) The largest contributor to total system variance is the Dryer inventory. Variance on the Dryer inventory is an order of magnitude greater than for any other process unit or transfer term. Dryer design should be reviewed with the objective of reducing holdup, facilitating cleanout, or maintaining a stable heel.
- (2) Material accumulation in SNM Storage should be minimized. Runout of stored SNM before materials balance closure is desirable because it will reduce storage variance.
- (3) Improved measurements in process units (other than the Dryer and SNM Storage) will not reduce total variance significantly. Therefore, major capital investment to improve these measurements is unwarranted. However, runout of unit inventories before materials balance closure may be an effective and economical way of reducing the variances associated with these units.

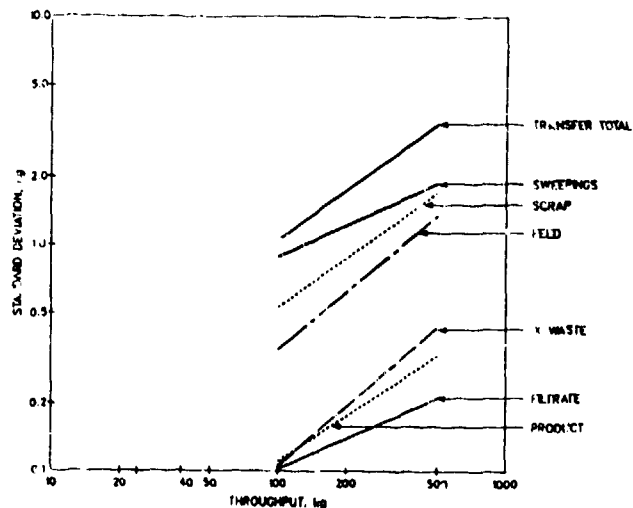


Fig. 3

Materials balance uncertainty for material transfers.

- (4) Tight physical protection, access control, and SNM control should be provided around the Dryer and SNM Storage because loss detection sensitivity in these areas is low relative to that of other process areas.
- (5) Improved SNM measurements for the Dryer and SNM Storage should be considered. Technology and cost limitations could limit the feasibility of this approach. Other approaches (equipment redesign, modified procedures, etc.) should be considered because they may be more practical and cost effective.
- (6) Breakup of the MBA into smaller unit process areas (at, before, and after Drying and SNM Storage) may not improve materials accounting. Materials accounting sensitivity could be decreased due to an inability to accurately measure cross MBA transfers.
- (7) Transfer variances do not contribute significantly to total system variance until process throughput exceeds 500 kg. Reduction of the materials balance closure period to less than 500 kg throughput intervals would reduce processing capacity and degrade loss detection sensitivity because thorough system cleanout may become impractical as cleanout frequency increases. At throughputs about 1000 kg, transfer terms become the controlling factor in total system variance.
- (8) The variance for Sweepings transfer is larger than that of the other higher throughput transfer terms. Improved methods for collection and measurement of Sweepings should be considered especially if materials balance closures occur at 500 kg or greater intervals.

#### IV. OPERATING BENEFITS

There are many operational benefits that can be realized from a good near-real-time materials accounting system. These operational pluses must be used to gain the cooperation and active support of Operations personnel. These positive operational features are described below and examples are given.

##### A. Material Loss Detection

Nuclear material processing operations frequently have sidestreams (typically wastes) that normally contain small to negligible amounts of product, valuable intermediates, or hazardous materials. These streams may not be monitored or sampled routinely because they normally contain small quantities of material or because the measurement is difficult, expensive, or impractical. Undetected, losses via these sidestreams can lead to an escape of valuable material and adversely impact operations because material must be recycled and reworked. Near-real-time materials accounting will allow the timely detection and determination of material loss via these sidestreams. The ability to quickly detect process upsets will improve processing efficiency and reduce the need for costly, capacity-stealing rework operations.

Los Alamos has detected and monitored loss of nuclear material using the materials accounting system when process instruments could not detect the loss and Operations personnel were unaware of this loss. In 1980-81, a series of tests were conducted at the Allied-General Nuclear Services, Barnwell Nuclear Fuels Plant, to test the concept of near-real-time-accounting (NRTA).<sup>1,2</sup> During these tests, a series of planned diversions were made to test the NRTA concept. At the completion of one test series, several diversions were detected by the NRTA system, and the point at which the material had been removed was pinpointed. However, one diversion was detected that was not a part of the exercise.

Figure 4 shows the system at Barnwell where this loss occurred. This pulse column system separates uranium from fission products. An aqueous stream containing dissolved uranium is fed to an Extraction column, where it is contacted by an organic stream containing an extracting agent. The uranium is extracted into the organic stream that exits the top of the column, whereas impurities remain with the aqueous stream that exits the bottom of the column. The uranium-bearing organic stream flows to a Stripping column where it is contacted with an acidic aqueous stream to recover the purified uranium.

Figure 5 shows a Cumulative Sum Materials Balance chart for the pulse column system. This chart plots the cumulative inventory discrepancy (ID) for hourly materials balance closure intervals. Planned diversions were made during the 20-40 hour and 80-90 hour time periods. The unplanned loss of material occurred during the 50-70 hour time period.

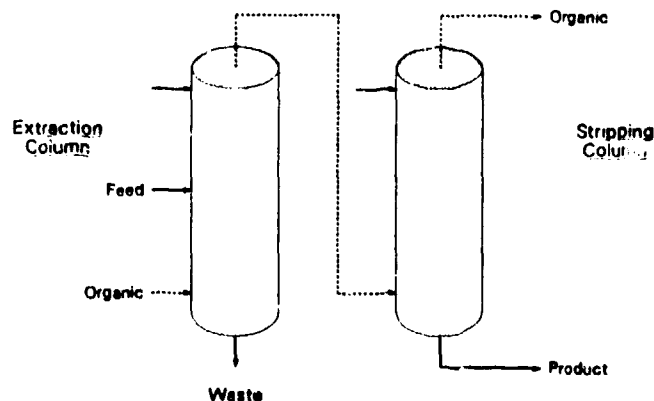


Fig. 4.  
Barnwell uranium recovery system.

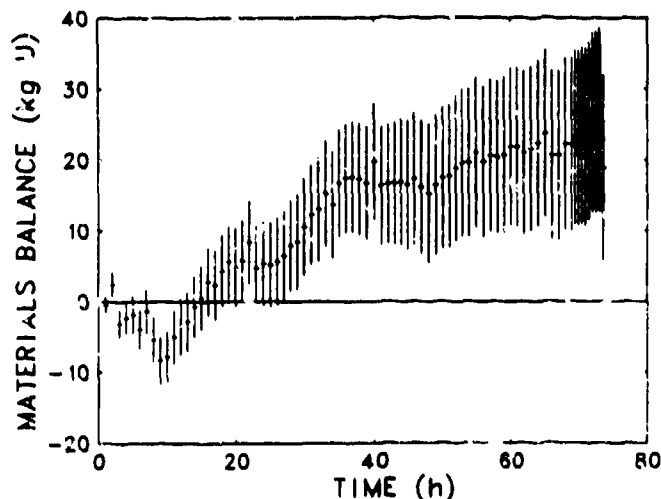


Fig. 5.  
Materials balance plots during uranium loss.

This diversion was identified by the accounting system as a loss of uranium to the Extraction column acid waste stream. Upon further checking, it was established that an unplanned removal of uranium had occurred at the time and location identified by the NRTA system because of an upset in the operation of the Extraction column. The normal process monitoring and control systems had not detected the upset conditions. If the accounting system had not been in place, a significant amount of material could have been lost requiring rework of the waste with a considerable loss of valuable material, lost processing time, and reduction in plant output while the recycle material was reworked.

This experience illustrates how a materials accounting system can be used to monitor material flows and losses where normal monitoring systems are either unavailable or their installation is impractical. This technique is especially useful where losses are suspected in streams that would

normally not be monitored. Ehinger has demonstrated the use of NRTA techniques to the monitoring of material losses and flows in experiments at Oak Ridge.<sup>2</sup>

The computerized materials accounting systems available in modern process plants allow closure of balances on individual transactions (material movements or process operations). Frequent closures on subMBAs or individual vessels will localize discrepancies leading to timely identification of process problems. This will facilitate timely adjustment of process parameters, maximizing process efficiency and minimizing costly rework operations.

#### B. Process Monitoring and Control

Process measurements must be utilized to realize a goal of comprehensive and complete materials accounting. Process instruments are typically less accurate and less well maintained than "accountability" instruments. Neutron monitors (provided for critical safety) on ion exchange columns, tanks, etc. are highly inaccurate and difficult to calibrate but can provide valuable safeguards information. An instrument of this type can be calibrated by using the MCCA system to track additions of material to individual process units. A materials accounting system that includes detailed materials tracking can monitor instrument performance, estimate measurement errors, and identify instruments that are out-of-calibration. Instrument biases can be followed and quantified as they develop, eliminating unknown errors and reducing instrument service frequency. Maintenance can then be scheduled only as needed.

Tank level measuring instruments are infrequently calibrated because of the time and effort required. The MCCA system can be used to check the calibrations; for example, when an intertank transfer is made, the quantity shipped and received should be checked not only to verify completion of the transfer but also to determine if the instruments are in agreement.

Figure 6 illustrates a three-tank system in which the second tank contains a level measuring device with a low bias. By comparing long-term data from these instruments, it is possible to locate and detect this bias. A materials balance drawn around the first two tanks would indicate that a continuous bleed of material (leak, diversion, etc.) is occurring. However, if the materials balance is redrawn to include the second and third tanks, it quickly becomes apparent that there is a problem with one of the measurements as there is now a net production of material that counterbalances the loss observed in the prior MBA. The amount of bias in the level measurement in the second tank can be quantified such that a temporary software fix can be used to recalibrate the system and correct prior measurement errors.

The MCCA system should incorporate software that compares related data to develop relative

errors, for example, comparison of volume received vs volume sent. This software can process and cross-compare relevant process data to develop the information necessary to establish and monitor the magnitude of process measurement errors.

Anomalies can frequently be resolved by re-drawing the MBA so that the missing (or suspect) data is not a critical component in the materials balance equation. In the above example, reformulation of the tank MBA to include all three tanks would have eliminated the false conclusions that resulted from the two-tank MBA analyses. This method is frequently effective in resolving anomalies caused by false readings, as the suspect instrument can be eliminated from the material's balance equation. A system of cross checks, alternate data sources, and consistency checks is mandatory if adverse impact upon operations due to bad data or other anomalies is to be minimized.

A modern MCCA system can quantify material losses to streams where measurements are of very poor quality or impractical. These data are used to quickly identify process upsets or misoperation of equipment or control systems. These lead to an improved understanding of the forces impacting process performance. The net result is an increase in both operating efficiency and production.

#### C. Holdup Evaluation

The ability to obtain a materials balance closure is generally not limited by inaccuracies of existing measurement but rather by unmeasured inventory or holdup. Spillage in glove boxes, heels in calciners, flourinators, deposits in vessels and pipes, etc. frequently control ID and its associated limit of error. Development of instruments to detect and quantify holdup can alleviate this problem. However, development of these instruments may be slow and expensive.

By the use of techniques similar to those used for checking instrument calibrations, it is sometimes possible to track the buildup of heels in vessels or spills in glove boxes. If supported by valid statistical methods, these data can be used to develop holdup estimators for the materials balance equation resulting in reduced IDs. Tracking of materials accumulation in small, localized process units during plant startup, cleanout, and restart can yield statistically supported estimators of unmeasured inventory that can be incorporated into the materials balance equation. This investment of time at startup can eliminate later shutdowns for system cleanout or anomaly resolution that would hinder plant productivity. Modern materials accounting methods can produce data that pinpoint material loss to holdup and quantify the magnitude and location of these apparent losses. These data will also show that what appears to be a significant loss is in reality the sum of many small material accumulations and measurement errors.



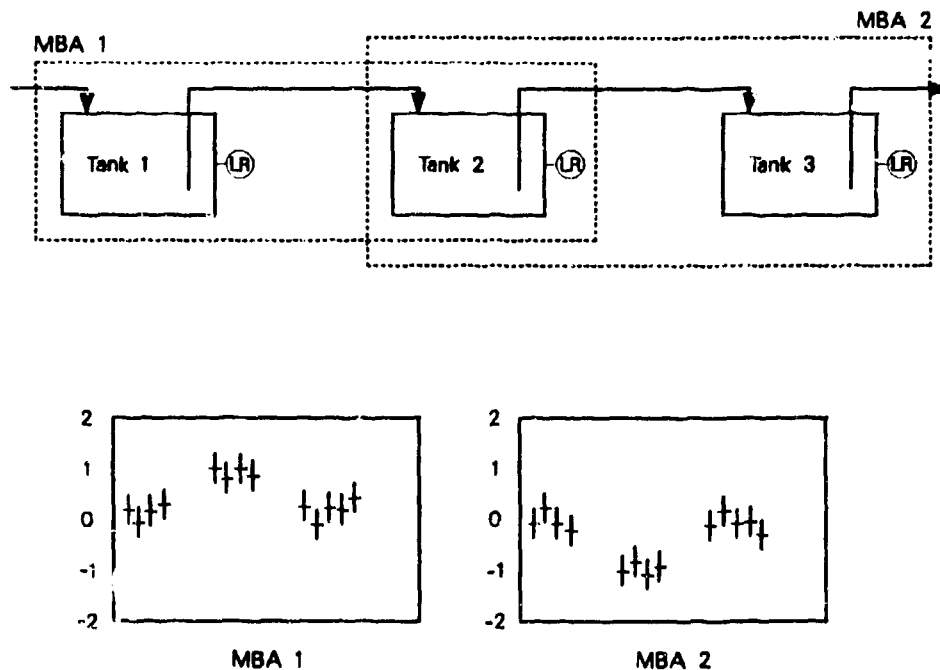


Fig. 6.  
Three tank systems with measurement error.

## V. CONCLUSIONS

The primary benefit of a materials accounting system to the plant operator is that it will help him to understand and follow the operation and performance of his plant. With data from the MC&A system, he can ascertain the impact of minor operating changes on performance. The process can thereby be fine-tuned to give optimum performance. Any upsets can be quickly detected and optimum conditions restored.

The MC&A system should not hinder plant operations and production. Operations personnel should use the MC&A system to provide an enhanced understanding of process system performance. MC&A personnel can provide assistance in the planning stages. During operation, the MC&A system can be used to detect process upset and calibrate process instrumentation. In summary, Operations personnel can use an MC&A system to their advantage rather than letting the MC&A system take advantage of them.

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